Supporting information

Kang et al.

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Materials and Methods

Well attribute estimation. For the measured abandoned wells, we used reports and databases from the Pennsylvania Department of Environmental Protection (DEP)[1] and the Pennsylvania Department of Conservation and Natural Resources (DCNR) [2] to estimate well depths (d), coal area designation (n_C, C, or C_1), plugging status (P), and well type (W) and to calculate the distances to the nearest active unconventional oil/gas well (r_U) and the nearest underground natural gas storage field (r_S) (Table S1). We do not consider well attributes such as well age, wellbore deviation, and abandonment method because of a lack of data for these attributes. The previous study that considered the largest number of well attributes used data from Alberta, Canada [3]. In Alberta, the Energy Resources Control Board (ERCB) has required testing, prior to abandonment, for surface casing vent flow (SCVF) across the province and for gas migration (GM) in a test area since 1995 [3]. Using the SCVF and GM results reported by operators and a database of well attributes, the researchers found that geographic area, wellbore deviation, well type, abandonment method, oil price, regulatory changes, and SCVF/GM testing all had a major impact on wellbore leakage [3]. Their study also showed that well age, well-operational mode, completion interval, and H_2S or CO_2 presence had no apparent impact on wellbore leakage [3]. However, the Alberta analysis explored the role of well attributes on the occurrence of SCVF/GM, not the magnitude of emission rates. In the U.K., elevated methane concentrations in the soil gas around "decommissioned" wells (plugged, capped, and buried) were found to occur within a decade of well decommissioning [4]. Although the U.K. study determined methane emissions rates, the study only considered age and geographic location (at the basin level) and did not include unplugged or not decommissioned wells, which are known to be prevalent in Pennsylvania and across the U.S. [5, 6, 7]. More recently, abandoned well measurements in four active production areas, Wyoming, Colorado, Utah, and Ohio, showed that plugging may be effective at reducing gas leakage [8]. Neither study considered plugged but vented wells.

We estimated d and W for the measured wells using the DCNR geospatial database [2] in *ArcGIS*. The DCNR database included 135,546 wells, 6,878 pools, and 676 fields. (Oil/gas fields consist of one or more oil/gas pools, from which oil/gas is produced.) The well dataset included both active and abandoned oil and gas wells. The pool dataset included the 2-dimensional outlines of geographical production limits for subsurface reservoirs containing oil, gas, or both. The field dataset included the 2-dimensional geographical boundary outlines of groups of pools related to a single stratigraphic or structural feature. Our analysis framework involved a comparison of field, pool, and well shapefiles to measured well locations using the near distance analysis function in *ArcGIS*.

For W, we first compared the types of the nearest/intersecting well, pool and field to the measured well in consideration. If the well/field/pool type (oil, including combined oil and gas, or gas) was consistent across datasets, W was assigned the corresponding type. If the well/field/pool type was not consistent across all three datasets, we considered the distance from the measured well to the nearest well, then the nearest and/or intersecting pool, and finally the nearest and/or intersecting field. If the nearest DCNR well was <50 m from the measured well, the DCNR well type was assigned to the measured well. We chose 50 m as the search radius based on our field experience of finding wells using a geographical coordinate system (e.g., GPS). (The accuracy of some DCNR well locations was uncertain because Pennsylvania's permitting requirements did not mandate precise survey location data until the mid-1980s [2].) If the nearest DCNR well was located >50 m from the measured well, the pool type was assigned if the measured well directly intersected the pool. Finally, when none of these approaches was successful, we considered the intersection of measured wells to fields, recognizing that multiple pools could be associated with a field and depths and other properties could vary within a field.

The depths of the nearest/intersecting field, pool, and well were used to assign d for each measured well. The pool depth was defined as the average of the producing formation depth provided in the DCNR database. The field depth was taken to be the depth of the pool with the largest number of wells within the field. The well depth was provided in DCNR's wells database. The depth of the measured well was taken to be the average of the nearest/intersecting well, pool, and field depths, unless: (1) there existed a well within 50 m of the measured well, (2) the nearest well depth was zero, or (3) the standard deviation of the three depths (well, pool, and field) was >1000 m. In 29 instances, the nearest well in the DCNR database was <50 m from the measured well. In these cases, the nearest well depth was assigned as the measured well depth. There were 20 nearest wells with depths of zero, presumably unknown, in which case the average depth of the nearest pool and field was used. Finally, if the standard deviation of the three depths (the nearest field, pool, and well depths) were >1000 m, one or two of the following three approaches were taken: (1) if the nearest well was >1000 m away, the well depth was omitted from the average; (2) if the measured well did not intersect a pool, the pool depth was omitted from the average; and (3) if the field displayed highly variable depths, the field depth was omitted.

We determined P of the measured wells based on the status specified in the DEP database, if available, and surface evidence (e.g., cementing or marker) [5]. The DEP database only indicates whether the wells are "plugged". Therefore, we are unable to differentiate between plugging techniques and other factors that may influence the effectiveness of the plug (e.g., casing integrity), which can vary, especially over time. We assumed wells that are "abandoned" or "orphaned" to be "unplugged". If the measured well could not be identified as matching a well on the DEP record (e.g., by the American Petroleum Institute number), we relied on field-based surface evidence to determine P. "Plugged/vented" wells were classified based on field investigation and were only found in coal areas as required in Pennsylvania. However, not all plugged wells that we determined to be in coal areas through our well attribute analysis were vented.

We assigned coal area designation by comparing well locations to mineable coal seams [9] and wells identified to be in coal areas by the DEP [1]. In Pennsylvania, a well is defined to be in a coal area if the well (1) overlies a mineable coal seam, (2) is ≤1000 ft (305 m) from the boundary of an area with a current Coal Mining Activity Permit, or (3) is in an area for which an underground coal mine permit application is under review [10]. We defined three different variables for coal area designation: n_C represented the number of mineable coals seams that the well intersects, C was an indicator of whether a well intersects any mineable coal seam, and C_1 was an indicator of whether a well intersects any mineable coal seams and/or was <50 m from a well identified to be in a coal area by the DEP. Coal seams were considered mineable if they were greater than 28 inches thick, greater than 100 feet from the surface, and laterally extensive [10]. Historical maps [9] showed coal seams that were located deeper than 100 feet from the surface and laterally extensive. These historical maps were manually digitized into *ArcGIS* shapefiles for coal seam areas greater than 28 inches thick. There were 13 maps, each showing different mineable coal seams occurring at different depths. We counted the number of coal seams that each measured well intersects based on locational information to obtain n_C. If $n_C > 0$, C was set as "coal" area, while if $n_C = 0$, C was set as "non-coal" area. Additionally, the DEP dataset of wells was used as another indicator for estimating whether a measured well was in a coal area. There were 31,676 wells in the DEP dataset and 6,740 wells were identified as coal wells. A near distance analysis was performed to determine how many coal wells in the DEP database were within 50 m of each measured well. If there were one or more DEP-designated coal wells within 50 m of a measured well and/or the $n_C > 0$, C_1 of the measured well was set as "coal" area. There were measured wells with no DEP wells within 50 m and thus, we could not rely on the DEP dataset alone to determine coal area designation.

For r_S, we determined the distance from the measured well to the closest point of the nearest underground natural gas storage field provided as shapefiles in the DCNR database [2]. For r_U, we computed the distance between the measured well and the nearest active unconventional oil/gas well in the DEP database [1].

Field measurements. We employed static chambers to measure methane flow rates at wells and control locations, following the methodology outlined in Ref. [5]. Uncertainties associated with the chamber measurements were discussed in Ref. [5]. In addition, we performed laboratory testing of the chambers at methane flow rates of 1 to 60 g/hr (in the order of high emitters) to study potential errors. We found that the chamber measurements underestimated the methane flow rate by an average of 13%. We adjusted the high emitters' methane flow rates for the final estimates (Figure 4) and the emission factors (Table 2).

Ten sampling campaigns were conducted from July 2013 to June 2015 (Table S2). The later campaigns were designed to broaden geographic coverage compared to earlier measurements and to obtain a representative sample based on well attributes, specifically P, C, and W (Table 1). The earlier sampling campaigns (July 2013 to January 2014) emphasized wells that were more easily accessible (e.g., closer to roads). For the later campaigns (March 2014 to June 2015), we prioritized well attributes over ease of access when selecting wells to measure. The objective was to have good estimates of emission factors for well categories identified as high emitters. Regions to focus our measurement efforts were determined using the DEP database and their oil and gas mapping tool [1]. We also repeated measurements at the same wells in McKean, Potter, Venango, Warren, and Lawrence Counties over multiple seasons.

Hydrocarbons. Methane, ethane, propane, and n-butane concentrations in field samples collected in 20 mL pre-evacuated glass vials were analyzed using gas chromatography (GC), as described in Ref. [5]. The methane concentrations were used to determine methane flow rates based on a linear regression of methane concentrations accumulated in a flux chamber over time [5, 11]. Slopes, upon which methane flow rates are based, with p-values greater than 0.2 were found to be essentially zero, or below detection level [5]. Ethane, propane, and n-butane concentrations were summed and presented with respect to methane concentrations (Figure 3).

Isotopes of methane. Carbon isotopes of methane for samples collected from March to October 2014 were analyzed using a cavity ring-down spectrometer (CRDS) at Princeton University, following the carbon isotope analysis procedure performed for July 2013 to January 2014 samples reported in Ref. [5]. The samples were collected in 125 mL pre-evacuated Wheaton™ glass flasks and 2 L SamplePro™ bags. We used a pure methane standard with $\delta^{13}C$ of -43.0‰±0.5‰ determined by Isotope Ratio Mass Spectrometry at the University of Toronto [12]. The standard was diluted to around 15 ppmv with Ultra Zero Air (Airgas, Inc.). The CRDS instrument determined the peak absorbance ratio of $^{13}CH_4$ over $^{12}CH_4$ for the standard methane and the samples. The pressure and temperature were maintained within ±0.1 torr and ±0.02°C respectively. The average time for each analysis was approximately 20 minutes, corresponding to the optimal integration time of the instrument. Between each sample, we flushed the sampling cavity with Ultra Zero Air and evacuated it three times. For ambient air methane concentrations, the precision of the $\delta^{13}C$ was estimated as ±2.0‰ (1σ) [12] based on the standard deviation of absorbance ratio of $^{13}CH_4$ over $^{12}CH_4$.

Carbon and hydrogen isotopes of methane for samples collected in 125 mL Wheaton™ glass flasks in January, March, and June 2015 were analyzed at Lawrence Berkeley National Laboratory. For CH_4 concentrations lower than ~1200 ppmv, carbon isotope ratios of CH_4 were measured by flushing 160 mL serum bottles into a Tracegas™ pre-concentrator interfaced with a Micromass JA Series Isoprime isotope ratio mass spectrometer (Micromass, Manchester, UK). Repeated injections of laboratory standards associated with sample analysis yielded a standard error ±0.4‰ (1σ; n=9). For CH_4 concentrations larger than ~1200 ppmv, hydrogen and carbon isotope ratios of CH_4 were determined separately using a gas chromatograph and an isotope ratio mass spectrometer interfaced with a pyrolysis reactor (GC-P-IRMS) and a combustion GC-C-IRMS, (Trace™ GC Ultra-Isolink™-DeltaV™ Plus system, Thermo Fisher Scientific, Bremen, Germany). CH_4 was separated chromatographically on an HP-molesieve fused silica capillary column (30 m x 0.320 mm). For hydrogen isotopes, after GC separation, CH_4 was pyrolyzed in an empty ceramic tube at 1450°C and the hydrogen isotope ratios were measured using the IRMS. For carbon isotopes, after GC separation, the CH_4 was combusted to CO_2 at 1000°C in a capillary ceramic tube and the carbon isotope ratio was measured in the IRMS. Repeated injections of CH_4 laboratory standard yielded a standard error of ±0.4‰ (1σ; n=10) for $\delta^{13}C$ and ±2.6‰ (1σ; n=14) for $\delta^{2}H$.

Carbon isotope ratios were reported in the conventional δ-notation relative to VPDB scale and hydrogen isotopes relative to SMOW.

Noble gases. Gas samples for noble gas analyses were collected in ~8" (inch) long, 1/4" diameter refrigeration-grade copper tubing. Before sampling, the copper tubes were flushed inline with at least 50 volumes of sample gas prior to sealing by manually pumping on the downstream side of the copper tube with a 3-way Luer Lock syringe. After purging, samples were sealed with either a 30/1000 of an inch gap stainless steel refrigeration clamp or CHA Industries (Fremont, CA) cold weld refrigeration crimper [13, 14].

In the laboratory, the gases were extracted from the copper tube on an ultra-high vacuum line (<2x10^{-9} torr) with an MKS 0-20 torr and 0-1000 torr capacitance manometers that are calibrated daily using a water-vapor free, inter-laboratory validated air standard collected on the shore of Lake Erie. The sample was collected at 12.1°C at a latitude of 41.5931 N. The laboratory standard was cross-validated between two independent laboratories (USGS and U.Rochester). Total gas pressure was measured and an aliquot (<10%) of total gas volume was pipetted into an SRS quadrupole mass spectrometer (MS) and an SRI GC, fitted with a flame ionization detector (FID) and thermal conductivity detector (TCD), for scanning and measurement of major gas and trace gas components (e.g., N_2, CH_4, O_2, H_2, Ar, CO_2, C_2H_6) [15, 16]. All samples were analyzed in triplicate. Standard analytical errors were all less than ±3.41% for major gas concentrations above the detection limit. The average external precision of "known-unknown" standards were reported in the parentheses (CH_4 (2.14%), C_2H_6 (2.78%), N_2 (1.25%), CO (3.41%), CO_2 (1.06%), H_2 (3.37%), O_2 (1.39%), Ar (0.59%)). These values were determined by measuring referenced and cross-validated laboratory standards including an atmospheric standard (Lake Erie Air) and a series of synthetic natural gas standards obtained from Praxair.

The isotopic analyses of noble gases were performed using a Thermo Fisher Helix SFT Noble Gas MS at The Ohio State University Noble Gas Laboratory following methods reported previously [15, 16, 17]. The average external precision based on "known-unknown" standards were all less than ±1.23% for noble gas concentrations with values reported in parentheses ([^{4}He] (0.89%), [^{22}Ne] (1.23%), and [^{40}Ar] (0.49%)). These values were determined by measuring referenced and cross-validated laboratory standards including an established atmospheric standard (Lake Erie Air) and a series of synthetic natural gas standards obtained from Praxair. Noble gas isotopic standard errors were approximately ±0.0095 times the ratio of air (or 1.27 x10^{-8}) for $^3He/^4He$ ratio, < ±0.681% and < ±0.814% for $^{20}Ne/^{22}Ne$ and $^{21}Ne/^{22}Ne$, respectively, and less than ±0.916% and 0.574% for $^{38}Ar/^{36}Ar$ and $^{40}Ar/^{36}Ar$, respectively (higher than typical because of interferences from C_3H_8 on mass=36 and 38).

Multilinear regression. We used multilinear regression to identify attributes that may predict methane flow rates ($\dot{m}$). Multilinear regression was performed using the *fitlm* function in *Matlab*. We considered numerous multilinear models but presented the results of six different models: L6a, L6b, L6c, L3, N6b, and N3. Model names have as their first letter an "L" if $\ln \dot{m}$ was used or "N" if $\dot{m}$ was used. The number indicates the number of variables in the multilinear regression model, where "6" means that six attribute variables are included. The last letter differentiates among models with the same number of variables and $\dot{m}$ representation.

Number of wells. We estimated the total number of wells drilled throughout the drilling history of Pennsylvania using all available data sources to obtain a range in possible well numbers. Because our goal was to determine the number of abandoned wells that may contribute to methane emissions, we considered all oil and gas wells drilled, including dry wells and those that were drilled for enhanced recovery. (Dry wells are wells incapable of producing enough oil or gas to justify completion. To enhance the recovery of oil, additional wells are drilled to inject fluids into the oil/gas bearing formation.) We did not include storage, test, observation, and waste disposal wells due to data limitations for years prior to 1957. For years after 1957, we assumed that these other well types are included in the modern digital records [18].

We compiled all available well numbers from data-based sources such as the published scientific literature [18], state databases [1], annual state reports [19, 20, 21, 22], and historical references detailing annual drilling and production [23, 24]. We did not include bulk estimates from references that did not provide estimation methods or annual data. For years for which reliable data were not available, we used relationships between oil production and the number of wells in the preceding or following 10-11 years. Historical oil production data were obtained from the 1993 Pennsylvania Department of Environmental Resources (DER) Progress Report [22].

Compiled annual well numbers were identified as including or excluding enhanced recovery wells. For years with well numbers that exclude enhanced recovery wells, we applied a factor based on the ratio of well numbers including and excluding enhanced recovery wells from other years to obtain our final estimate of well numbers.

To estimate the total number of abandoned wells, including those that have been orphaned and/or lost, we took the total number of wells drilled and subtracted from that total the number of active and inactive oil/gas wells in Pennsylvania as of March 1, 2014, which is 10,921. Our total number of abandoned wells represents wells drilled and abandoned, with and without plugging, from 1859 to 2013.

Methane emissions. We estimated the state-wide methane emissions from abandoned oil and gas wells in Pennsylvania using an attribute-based approach and our estimates of well numbers. We used the DEP database [1] to determine the proportion of wells with all combinations of W, C, and P (Table 2) and then scaled the numbers by the total estimated number of abandoned wells. There were 12 well types (including "undetermined"), which we aggregated into "oil", "gas", "other", and "undetermined". Our "oil" wells designation included "oil" wells and "combined oil & gas" wells. Our "gas" wells designation included "gas" wells and "coalbed methane" wells. "Other" wells were "injection", "dry holes", "storage", "observation", "multiple well bore type", "waste disposal", and "test" wells, to which we assigned the general average emission factor (Table 2). For "undetermined" wells and wells not assigned a type, we applied a weighted emission factor based on the distribution of well types in the DEP database [1]. For coal area designation and plugging status, we used the emission factors in Table 2 directly.

Measurement Data and Well Attributes

R^2 of methane flow rates at wells. Methane flow rates from abandoned oil and gas wells, $\dot{m}$ (mg hr^{-1} well^{-1}), were based on linear fits of methane concentration accumulation over time [5]. The R^2 values for these linear fits showed that improved fits were more easily achieved for higher flow rates (Figure S1). Most methane flow rates had R^2 values greater than 0.8. The distribution in R^2 values remained similar for measurements conducted in all three years: 2013, 2014, and 2015.

Variation in methane flow rates at wells. Variances (and the coefficient of variation) in methane flow rates at wells were not related to the number of measurements at a given well site (Figure S2). Therefore, the relationship between variances and methane flow rates discussed in the main text (Figure 2) were not affected by the number of repeat measurements.

Methane flow rates at control locations. Methane flow rates at control locations (Figure S3) were generally orders of magnitude lower than those found at wells, similar to observations reported in Ref. [5]. Although methane flow rates could be similar to control flow rates at the lower end, control flow rates did not go above 100 mg hr^{-1} location^{-1} while methane flow rates at abandoned wells frequently exceeded 100 mg hr^{-1} well^{-1} and reached as high as 10^5 mg hr^{-1} well^{-1}. The mean flow rate at controls was 0.6 mg hr^{-1} location^{-1}. Of the 193 control location measurements, 91 measurements (47%) showed methane emissions below detection level (i.e., no methane accumulation or update based on the p-value). There was a similar number of negative methane flow rates (i.e., methane sinks) (53 measurements or 27% of the control measurements) as positive methane flow rates (i.e., methane emissions to the atmosphere) (49 measurements or 25% of the control measurements).

Noble gas measurements at wells. The noble gas (e.g., ^{4}He) and methane concentrations were evaluated with ^{22}Ne and ^{36}Ar as environmental tracers of air and/or air-saturated water (Figures 3 and S4). High methane-emitting gas wells had $^3He/^4He<0.10R_A$ (where R/R_A is the ratio of ^{3}He to ^{4}He in a sample compared to the ratio of those isotopes in air and R_A nomenclature denotes the $^3He/^4He$ ratios of samples with respect to air), $^4He/^{22}Ne>100$, $[^4He]>10^{-4}$ ccSTP/cc, and $CH_4/^{36}Ar> 5 \times 10^3$. Selecting ratios of key gas parameters to ^{22}Ne and ^{36}Ar provided a comparison between deep, thermogenic, crustal sources (^{4}He and CH_4) vs. atmospheric/air-saturated water (^{22}Ne and ^{36}Ar). Because the atmosphere is extremely well mixed, ^{22}Ne and ^{36}Ar in air and air-saturated water are ubiquitous on the Earth's surface and in meteoric water. ^{22}Ne and ^{36}Ar are nearly constant in air and in meteoric water, and are very well constrained. As a result, minor variations in the $^4He/^{22}Ne$ or $CH_4/^{36}Ar$ ratios readily record minor contributions from deep crustal fluids at the surface. These ratios are specifically sensitive to gas leakage from natural gas wells in Pennsylvania, which contain relatively dry natural gas (i.e., low C_{2-4}/C_1). In hydrocarbon fluids that have achieved lower relative thermal maturities throughout their geological histories (i.e., fluids that contain more wet (C_2-C_4) gases and/or oil-associated gases), there is typically lower gas-water ratios (e.g., $^4He/^{22}Ne$, $^4He/^{36}Ar$, or $CH_4/^{36}Ar$) [13, 14, 15]. Hence, wet and oil-associated methane typically has progressively lower $^4He/^{22}Ne$, $CH_4/^{36}Ar$, or $^4He/^{36}Ar$ than non-oil associated dry natural gas. For these reasons, the $^4He/^{22}Ne$, $CH_4/^{36}Ar$, or $^4He/^{36}Ar$ was relatively high in natural gas wells as opposed to oil-associated gases. The relative increases in $^4He/^{22}Ne$ (and $CH_4/^{36}Ar$) effectively served as a proxy for the original thermal maturity at which hydrocarbon gases were produced in the source rocks [13, 14, 15]. Therefore, $^3He/^4He$, $^4He/^{22}Ne$, and $CH_4/^{36}Ar$ were able to differentiate between gas and oil or combined oil & gas wells.

Noble gases did not differentiate between plugging statuses or coal area designation.

The impact of d, n_C, r_U, and r_S on methane flow rates. Here, we evaluated the role of d, n_C, r_U, and r_S on $\dot{m}$ (Figure S5), which were not presented in detail in the main text. Visually, there appeared to be a relationship between d and $\dot{m}$. However, much of the dependence of $\dot{m}$ on d could be explained by the dependence of $\dot{m}$ on W, as $\dot{m} > 10^3$ mg hr^{-1} well^{-1} were dominated by gas wells. Gas wells were drilled to both shallow and deep depths, whereas oil wells tended to be relatively shallow. For wells in coal areas, $\dot{m}$ appeared to increase with the number of intersecting workable coal seams (n_C). However, several high emitters had no intersecting workable coal seams, suggesting that other factors were also important. Finally, high methane-emitting wells were up to $\sim$20 km away from the nearest active unconventional well and the nearest underground natural gas storage field, more than the average distance of all wells (18 km for storage fields and 8 km for unconventional wells).

Multilinear regression. Multilinear regression analysis of six different models showed that Model L6b was the best predictor of $\dot{m}$ (Table S3). However, the p-values of the coefficients for d, r_S, and r_U were above 0.05 and these terms did not play a significant role in determining $\dot{m}$. In fact, Model L3 was able to provide a similar fit to Model L6b. The two models, N6b and N3, that were not based on the logarithmic values of $\dot{m}$ performed significantly worse than the models based on $\ln \dot{m}$.

For upscaling methane emissions, it is important to get the correct order of magnitude of the methane flow rates of high emitters. None of the models was able to reproduce $\dot{m}$ at the order of magnitude level (Figure S6). Therefore, we did not use the models to estimate methane emissions.

The multilinear regression analysis showed that W, C, and P were the best predictors of $\dot{m}$. For coal area designation, we used C instead of n_C and C_1, both of which had p-values greater than 0.05 (Figure S6).

Estimate of Well Numbers

A brief history of oil and gas development in Pennsylvania. Pennsylvania has the longest history of oil and gas production in the U.S. The first commercial oil well in the U.S., the "Drake Well", was drilled in 1859 in Titusville, Pennsylvania. In 1881, the Bradford Oil Field in northwest Pennsylvania produced 83% of America's oil output [25]. After production in the Bradford Oil Field peaked in 1881, the field continued to produce significant quantities of oil using enhanced, or "secondary", recovery methods, mainly involving waterflooding [26].

Waterflooding involves drilling additional wells to inject water in oil formations and increase the flow of oil to producing wells [27]. It differs from primary oil production that uses natural pressures or pumping without any injection wells. These enhanced recovery (ER) wells, which are not producing oil or gas wells, can also act as conduits for subsurface fluid migration and gas emissions at the surface.

The large potential number of ER wells and the lack of historical reporting of these wells make them both important and challenging to quantify. The first use of water flooding occurred in 1880 in the Venango Oil Field in Pennsylvania [28]. Water flooding was illegal in Pennsylvania until 1921 [26] and water flooding wells drilled prior to 1921 were unlikely to have been reported or recorded. Even after 1921, ER wells may not have be considered as oil and gas wells and we could not find records of injection wells until 1950, when Pennsylvania's Department of Environmental Resources (DER) began publishing progress reports for oil and gas development in the state [19]. The "five-spot" method, a popular water flooding technique developed in 1927, involves drilling four additional injection wells for each producing oil well [26, 28]. Another popular method used was the "seven-spot" pattern with six injection wells per producing well [26]. Because producing and injection wells are likely to be in a grid pattern, there will likely be one additional row of injection wells per row of producing wells for the five-spot method. Therefore, inclusion of injection wells will increase the estimated number of wells by a factor of at least two.

Substantial secondary oil production, mainly through waterflooding, occurred in Pennsylvania in the 1930s and 1940s [22]. Oil production and enhanced oil recovery in Pennsylvania decreased steadily after the 1930s/1940s peak [22].

Natural gas production in Pennsylvania began in the late 1800s. Between 1882 and 1928, Pennsylvania's natural gas production was the second highest in the Appalachian Basin, after West Virginia [24]. In the early 1950s, discoveries of conventional natural gas reserves in deep gas fields led to growth in natural gas production [21]. More recently, Pennsylvania has experienced significant growth in natural gas production attributable to horizontal drilling and hydraulic fracturing of shale formations (e.g., the Marcellus formation) and has been the focus of scientific and media attention from both economic and environmental standpoints [6, 29].

Compilation of data sources. Information on the number of wells drilled from 1859 to 2013 requires compilation of different data sources, each covering different time periods (Table S4 and Figure 5). For 1859-1929, we obtained numbers of wells drilled from two historical books on oil and gas production [23, 24]. For 1930-1949, we estimated well numbers based on oil production and trends in preceding and following years. For the 1950-1991 time period, the Pennsylvania DER published annual to bi-annual progress reports on oil and gas development [19, 20, 21, 22]. (The discontinuation of these reports is likely due to the split of DER into the DCNR and the DEP in 1995.) The DCNR now manages a digital database known as the Pennsylvania Internet Record Imaging System/Well Information System (PA*IRIS/WIS), which is being modernized and renamed as EDWIN (Exploration and Development Well Information Network). This database was used to estimate well numbers in Ref. [18]. The PA*IRIS/WIS and EDWIN database of wells were different from the DCNR wells dataset [2] used in our attribute estimation framework. The DEP also maintains data on wells drilled, which was publicly available on their website [1]. In addition, since 2009, the number of wells drilled as reported by operators to the DEP was also available [30].

The total number of wells drilled in Pennsylvania from the start of drilling in 1859 until 1928 was reported to be 168,190 [24], which corresponded to an average of 2403 wells per year. Figure 5 showed the number of wells drilled per year from 1889 to 1920 in Pennsylvania [23] and from 1859 to 1928 in the Pennsylvania and New York (NY) portion of the Appalachian Basin [24]. During this time period, 89% of the total production was in Pennsylvania with the New York portion of the Appalachian Basin becoming only marginally significant in 1876 [24]. The two historical sources [23, 24] show that the first peak in well drilling occurred in the 1890s when approximately 6000 to 7000 wells were drilled annually. There was no mention of ER well counts in either of these historical references.

We estimated the number of wells drilled from 1929 to 1949, excluding ER wells drilled before 1938, to be 68,000.

For 1929, we estimated a well number of 3600, which was determined by scaling the combined NY and PA well number of 4009 by 0.89 (see previous paragraph). For most years from 1930 to 1976, well numbers were available from Oil Weekly annual activity reports [18], Minerals Yearbooks by the U.S. Geological Survey [18], and PA DER Progress Reports [19, 20, 21, 22]. No data for all of Pennsylvania were available for 1931, 1932, 1936, 1947, 1950, and 1953 and previous estimates used linear interpolation of wells numbers from the preceding and following years [18]. The well numbers given in Ref. [18] for 1930 to 1949 appeared to be an underestimate since the numbers conflict with other data sources and trends. For one, the well numbers in Ref. [18] for all of Pennsylvania were lower than those reported for the Bradford Field alone [26]. In addition, there was a drop in well numbers in Ref. [18] in a period of increasing production (1930-1937). Analysis of well numbers and oil production data from other time periods showed that well numbers generally increased with production (Figure S7). Data from 25 water flooding projects in northern Pennsylvania that began between 1927 and 1946 showed that an additional 1.5 "water-intake" wells were drilled for every new producing well drilled [26]. Therefore, the low numbers in Ref. [18] could not be explained by assuming that most of the additional wells were injection wells, not producing wells. Because the well numbers given in Ref. [18] appeared to be incomplete and inconsistent with our understanding of the oil and gas history in the region, we used oil production data to estimate well numbers across Pennsylvania for 1930 to 1949.

The oil production-well numbers relationship was dependent on whether production was increasing or decreasing. Oil production increased significantly from less than 8 million barrels in 1920 to 19 million barrels in 1937 (Figure 5) [22]. In the years preceding 1930, we saw a linear relationship between oil production and well numbers ($R^2 = 0.63$) as oil production increased (Figure S7). Using this relationship, the number of wells drilled in 1930 to 1937 was estimated to be 32,000. Oil production began to decrease in 1938. Data from the Pennsylvania DER Progress Reports also showed a linear relationship between oil production and well numbers ($R^2 = 0.81$) for 1950 to 1959, a period of decreasing oil production. The number of wells for 1938-1949 using this second relationship was 32,000.

The DCNR's PA*IRIS/WIS data produced a number for well completions of 114,154 for 1957 to 2012 [18] (Table S4). The DCNR well number, which includes ER wells, was assumed to be accurate because previous research found the database to be the most "complete and internally consistent digital data record of documented wells in Pennsylvania" [18].

To consider the role of ER wells, we reviewed well numbers from the Pennsylvania DER Progress Reports and the DEP database. The DEP database contained 1738 "injection" wells, which represented 5% of the wells on the database. In contrast, the total number of wells drilled from 1950 to 1991 based on the Pennsylvania DER Progress Reports was 55,516 excluding ER wells and 65,286 including ER wells, which corresponded to 18% of the wells being ER wells. The annual DER well numbers including and excluding ER wells showed that the relative proportion of wells drilled for ER was the highest in the early 1950s. In 1951, the inclusion of ER wells increased the number of wells by a factor of 4 (Producing & Injection Wells / Producing Well). Using the 1950 to 1952 data, we calculated ratios of total wells including ER wells to producing wells (excluding ER wells) and obtained an average of 3.5. These ratios were in line with data from 25 water flooding projects in the Bradford Field, which have factors ranging from 1.7 to 3.3 with a mean of 2.5 [26]. We used the total well numbers including both oil and gas for these ratios because we did not have a reliable breakdown of oil and gas wells for the 1930-1937 time period.

Drilled well numbers were also available on the DEP's website beginning from 1940 (Figure 5). However, the DEP's well numbers prior to 1956 ranged from 1 to 22. These low numbers were inconsistent with our understanding of oil and gas development in Pennsylvania and we did not use the DEP numbers in our estimates. We used the DEP numbers for 2013 only because Ref. [18] did not provide well numbers after 2012.

For most years with data available, the DER and DEP numbers were underestimates when compared to the DCNR numbers (Figure 5). Nonetheless, the general trends in all three sources were similar. In all three data sources, there was a peak in the number of wells of up to 6500 wells drilled per year in the 1980s, which was followed by a steep decline to ~1000-2000 wells drilled per year by the 1990s. The DEP and DCNR numbers also showed a 2007 peak in the number of wells drilled in Pennsylvania of ~5000-6200 wells drilled per year. By 2012, the number of wells drilled per year dropped to ~2000-3000.

Uncertainties. Modern digital records managed by state agencies are known to have poor records of wells drilled before 1957 [18]. We compared the DCNR numbers, which were assumed to be correct for 1957 onwards [18], to two historical data sources: the Pennsylvania DER Progress reports and the Minerals Yearbook. Considering data from 1957 to 1976, we found that the DCNR numbers were 1.3 times larger than the numbers from the PA DER Progress reports and 1.6 times larger than the number from the Minerals Yearbook. For years that we used historical data sources (i.e., the DER Progress Reports or Ref. [24]), we conservatively used a factor of 1.3 to account for underestimation due to missing data and other uncertainties for all years before 1957 (Table S4). Overall, we estimated ~150,000 wells as unaccounted for due to underreporting and lack of documentation (Table S4).

A comparison of the annual drilled well numbers showed that there are discrepancies between data sources even for recent years. For 1992-2012, the DCNR numbers were on average 1.5 times larger than the DEP numbers. For 2009-2013, the number of wells reported by operators and the number of wells on the DEP's website were similar but not equal. Therefore, for 2013, a year for which there are no DCNR numbers, we used a factor of 1.5 times the DEP number to estimate the upper limit in the well number.

Another major source of uncertainty was the inconsistency in terminology. Well numbers given in Ref. [18] including oil, gas, and dry wells were stated to be the number of well completions. However, dry wells are typically not completed. It was unclear if the wrong terminology was used or if they represented some subset of dry wells that were completed. Here, we assumed the former and that all dry wells, both completed and not completed, were included in the well numbers given in Ref. [18]. However, this assumption may lead to an underestimate in our 1957-2012 well numbers.

Although we used data-based methods where possible, uncertainty remains in extrapolating data from different time periods to periods without data. This applied both to factors used to represent ER wells and underreporting.

Well numbers for years prior to 1957 may not have included oil/gas wells such as observation and test wells. Available historical data sources specifically stated numbers for oil, gas, and dry wells and made no mention of other oil/gas well types [24].

Total number of wells. Based on the trends and the history of oil and gas in Pennsylvania, we assumed that ER via waterflooding played a significant role in the years prior to the 1950s. We scaled the well numbers for 1859 to 1929 by 1.5 or 2.0 and the well numbers from 1930 to 1937 by 2.0 or 3.5 (Table S4). Given the uncertainty in the number of potential ER wells before 1921, we reduced the factor of 2.0 applicable for the five-spot method to a factor of 1.5 for 1859-1928 to obtain a lower bound estimate. The bulk of the wells in Pennsylvania were drilled after 1875 (Figure 5), shortly before the first ER wells were likely to be drilled. Therefore, a factor of 2.0 may also be reasonable for 1859-1928, and this factor was used as the upper bound estimate for well numbers including ER wells. For the upper bound number for 1930-1937, we used a factor of 3.5 determined using the average of well numbers from the Pennsylvania DER annual reports for 1950-1952, when ER activities were more likely to resemble earlier periods. From 1938 onwards, we directly used the well numbers based on the DER progress reports or the DCNR well numbers [18] as they included ER wells. The total number of ER wells that were previously unaccounted for was estimated to be 110,000 to 250,000.

Adding ER wells and accounting for potential underreporting, we calculated the number of abandoned wells to be 470,000 to 750,000 for the state of Pennsylvania (Table S4). These numbers also included dry wells for all years and other well types (e.g., observation and test wells) for 1957 onwards.

Number of wells by attribute. It is important to estimate not only the total well numbers but also the numbers by attribute, especially for the three factors identified to be important predictors of high methane-emitting wells (well type, plugging status, and coal area designation).

We compiled historical data from various sources to estimate the proportion of wells that were oil and gas (Table S5). Considering only oil and gas wells (not dry, test, or other wells) to determine proportions, we found that oil wells may represent 65% to 76% of abandoned wells; while gas wells may represent 24% to 35% of abandoned wells. In contrast, the DEP database showed a breakdown of 50% oil and 50% gas wells [1].

Unfortunately, historical data to estimate the number of wells by plugging status or coal area designation were not available. In the DEP database, 70% of abandoned wells were plugged, leaving 30% unplugged [1]. Based on the long history of oil/gas development in Pennsylvania and poor historical records, the actual number of unplugged wells was likely to be higher. As for coal area designation, the DEP database showed that 21% of wells were in coal areas. Of these wells in coal areas, 86% were plugged and could be assumed to be plugged/vented.

Methane Emission Estimates

Methane emissions from abandoned oil and gas wells was estimated to be 0.040 to 0.066 Mt (10^{12} g) CH_4 per year in Pennsylvania using well numbers of 470,000 to 750,000 (Table S4). These emissions represented 5% to 8% of total anthropogenic methane emissions for Pennsylvania in 2011, which was estimated by the World Resources Institute (WRI) [31] to be 15.26 Mt CO_2e per year (0.73 Mt CH_4 per year). WRI used a global warming potential (GWP) of 21 following the second assessment report of the Intergovernmental Panel on Climate Change and used the State Inventory Tool (SIT) of the U.S. Environmental Protection Agency (EPA) [31]. The use of the GWP of 21 does not impact our percentages since they are in terms of mass of methane. The source categories included in the WRI estimates were energy, agriculture, industrial processes, waste, land use and forest, and bunker fuels. The WRI estimates contained uncertainties and may have underestimated total state-wide GHG emissions (including CO_2) by a few Mt CO_2e per year [31]. Furthermore, there were year-to-year variabilities. Considering the period from 2001 to 2011, the minimum and maximum methane emission estimates were 13.1 and 17.6 Mt CO_2e per year [31].

The above methane emission estimates were based on the distribution of attributes in the DEP database, which may not be representative of actual distributions of all abandoned wells in Pennsylvania. Nonetheless, it was the only source that provided a breakdown of wells by attributes (including well type, plugging status, and coal area designation). Although we could estimate the number of oil vs. gas wells, we still needed to rely on the DEP database for the proportion of plugged wells and the proportion of wells in coal areas. If we scaled the well numbers by the highest percentage of oil wells estimated using historical data (our Estimate 3 in Table S5), methane emissions from abandoned oil and gas wells went down to 0.02 to 0.04 Mt CH_4 per year in Pennsylvania, which corresponded to 3% to 5% of total anthropogenic methane emissions in 2011 for the state. However, this estimate assumed that the distribution of plugging status and coal area designation in the DEP database was correct. A change in these distributions could both increase and decrease the total methane emissions from abandoned wells. For example, increasing the percentage of wells in coal areas from 21% to 31% increased methane emissions to 0.05 to 0.08 Mt CH_4 per year in Pennsylvania, which corresponded to 7% to 12% of total anthropogenic methane emissions in 2011 for the state. Overall, more studies are needed to better estimate the distribution of plugged wells and wells in coal areas to further improve methane emission estimates.

Uncertainties in the methane emission estimates could be addressed with additional data, including information on estimate emission factors, well attributes, and well numbers. For example, field data obtained using geophysical methods could be used to improve estimates of the depths of measured wells, in addition to assessing plugging and casing conditions. Production or other well data that were not publicly available may be collected from industry and used to estimate the well attributes, which could then be used to verify the well attribute estimation approach. Well-finding methods including magnetometry surveys and field visits could be used to estimate errors in well numbers. Finally, additional field measurements of abandoned wells with various well attributes, especially undersampled categories such as plugged oil wells in coal areas and unplugged gas wells in noncoal areas (Table 2), could improve emission factors. These data collection and analysis efforts are needed not just in Pennsylvania, but also in the many other states (e.g., West Virginia, Texas, and California) and other countries with a long history of oil and gas development.

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Table S1. Well attributes

Attribute	Type	Variable
Depth	Continuous	d
Coal Area Designation Options		
Number of Intersecting Mineable Coal Seams	Continuous	n_C
Coal Indicator*	Categorical (2)	C
Alternate Coal Indicator**	Categorical (2)	C_1
Plugging	Categorical (3)	P
Well Type	Categorical (2)	W
Distance to Nearest Underground Natural Gas Storage Field	Continuous	r_U
Distance to Nearest Active Unconventional Well	Continuous	r_S

The number in parentheses for categorial variables indicates the number of categories.
* Intersection with one or more mineable coal seams [9].
** Intersection with one or more mineable coal seams [9] and within 50 m of a well designated to be in a coal area by Pennsylvania's DEP [1].

Table S2. Sampling rounds

Sampling Campaigns		Number of Well	
Year	Month	Measurements	Counties
2013	July-August	14	McKean
2013	October	13	McKean
2014	January	14	McKean, Potter
2014	March	11	McKean, Potter
2014	June	15	McKean, Potter
2014	July	17	Venango, Lawrence, Allgheny
2014	October	22	McKean, Potter, Venango, Lawrence
2015	January	4	McKean, Potter
2015	March	26	McKean, Potter, Warren
2015	June	27	McKean, Clearfield, Venango, Warren

Table S3. Multilinear regression analysis results: R^2 values, p-values, and variable coefficients.

	Model L6a	Model L6b	Model L6c	Model L3	Model N6b	Model N3
R^2 for model	0.39	0.44	0.26	0.43	0.20	0.20
p-value for model	8.1×10^{-7}	4.4×10^{-8}	8.0×10^{-4}	1.9×10^{-9}	0.011	0.0010
Variable Coefficients						
Intercept	2.54	2.84*	2.49	3.23***	18317	17159
d	0.00049	0.00039	-9.41×10^{-5}		-0.39	
$C =$ coal area		-5.50***		-4.95***	-17384	-16486
$C_1 =$ coal area			-1.12			
n_C	-1.39***					
$P =$ unplugged	3.58***	3.99***	3.40**	3.94***	24107*	24711*
$P =$ plugged/vented	9.60***	8.33***	7.00*	9.85***	67777*	67842**
$W =$ Oil	-3.33*	-2.88*	-3.72*	-3.35***	-34302	-29930**
r_S	0.037	0.016	0.015		286	
r_U	-0.095	-0.087	0.053		-290	

Models L6a, L6b, L6c, and L3 are based on $\log \dot{m}$; while Models N6b and N3 are based on $\dot{m}$.
* p-values<0.05.
** p-values<0.01.
*** p-values<0.001.

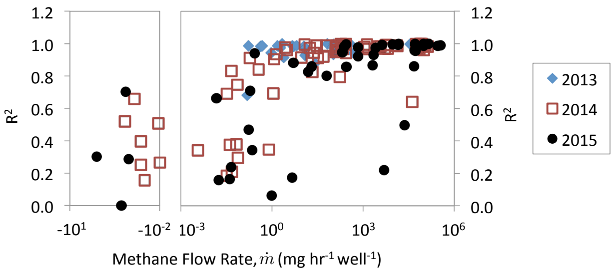

Fig. S1. R^2 values of methane flow rate measurements ($\dot{m}$).

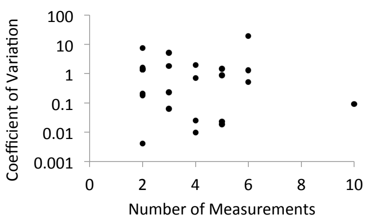

Fig. S2. Coefficient of variation vs. number of measurements at 27 well sites.

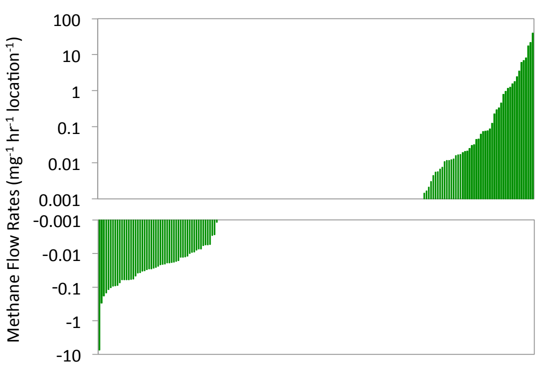

Fig. S3. Methane flow rates measured at 193 control locations, which are generally <10 m from a measured well site. Each bar represents a methane flow rate measurement, of which 53 are negative and 49 are positive. The large region of the plot with no bars represents the 91 measurements with flow rates below detection level.

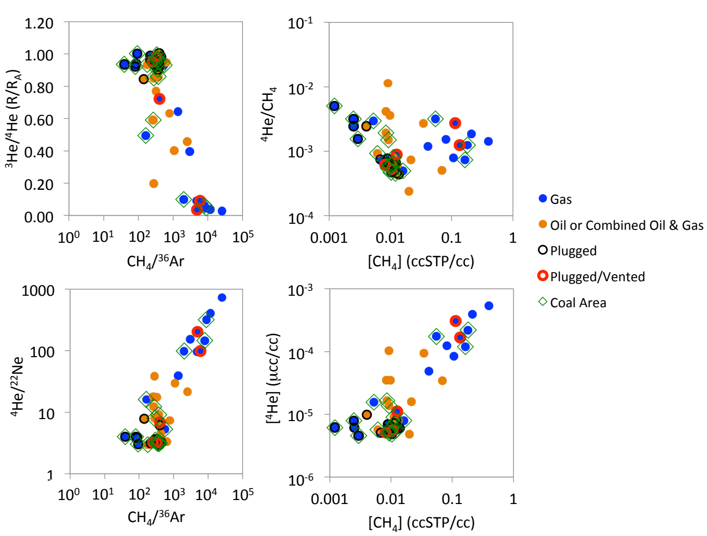

Fig. S4. Noble gas data with respect to C, W, and P. For repeat measurements, the average of the data for the well are shown.

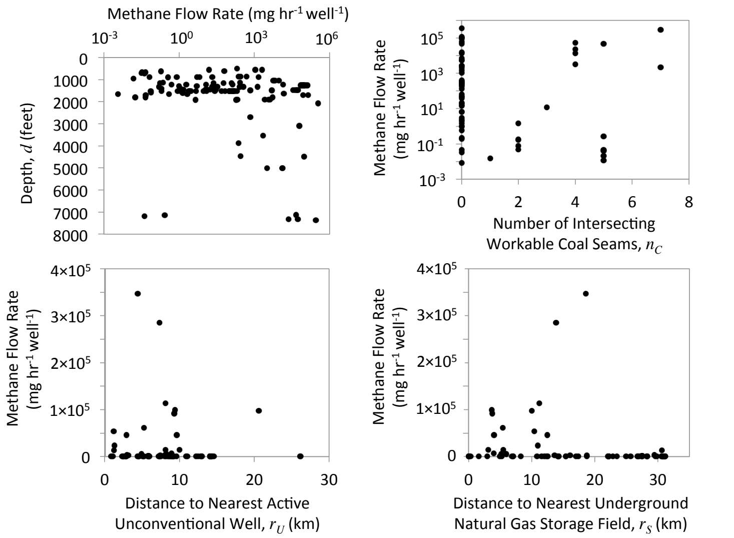

Fig. S5. Methane flow rates ($\dot{m}$) vs. depth (d), number of intersecting workable coal seams (n_C)[9], distance to active unconventional oil and gas wells (r_U), and distance to underground natural gas storage fields (r_S).

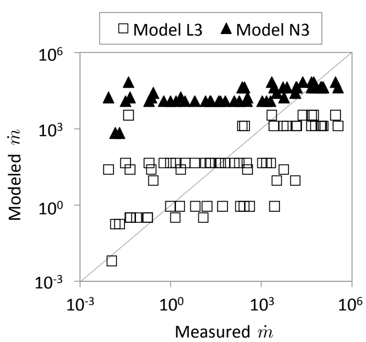

Fig. S6. Modeled and measured methane flow rates, $\dot{m}$ (mg hr^{-1} well^{-1}).

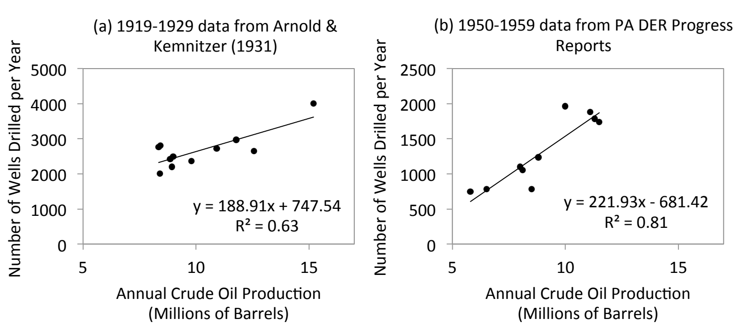

Fig. S7. Oil production [22] and well numbers from two historical sources.

Table S4. Number of oil/gas wells in Pennsylvania including enhanced recovery (ER) wells and potential underreporting.

				Incl. ER Wells*				Underreporting**	
Year(s)	Number	Source	Description	Factor†	Number	Factor†	Number	Factor†	Number
1859-1928	168,190	Table 12, Arnold & Kemnitzer [24]	No ER wells	1.5	250,000	2	340,000	1.3	440,000
1929	3600	Scaled for PA from Table 13, Arnold & Kemnitzer [24]	No ER wells	2	7200	2	7200	1.3	9300
1930-1937	32,000	Model 1919-1929 based on Arnold & Kemnitzer [24] (Figure S7)	No ER wells	2	64,000	3.5	110,000	1.3	140,000
1938-1949	32,000	Model 1950-1959 based on data from PA DER Progress Reports [22] (Figure S7)	Incl. ER wells	1	32,000	1	32,000	1.3	46,000
1950-1956	10,443	PA DER Progress Reports	Incl. ER wells	1	10,443	1	10,443	1.3	14,000
1957-2012	114,154	Modern Digital Records (PA*IRIS/WIS), Dilmore et al. [18]	Incl. ER wells; Completed wells only (but incl. dry)	1	114,154	1	114,154	1	114,154
2013	2092	PA DEP Oil and Gas Reports [1]	Incl. ER wells	1	2092	1	2092	1.5	3000
TOTAL 1859-2013	360,000				480,000		610,000		760,000
Active and Inactive Wells (3/1/2014)	10,921				10,921		10,921		10,921
ER Wells 1859-1937	0				110,000		250,000		250,000
Additional Wells due to Uncertainties** 1859-1956	0				0		0		150,000
TOTAL ABANDONED 1859-2013	350,000				470,000		600,000		750,000

Our estimates are rounded to two significant digits. Numbers from other sources are not rounded.
† Factors represent (number of producing & injection wells) / (producing wells).
* We multiply well numbers that do not include ER wells by the specified factor, determined based on data and/or the assumption that the five-spot method is used.
** We multiply well numbers including ER wells by the specified factor, determined based on comparison of historical and modern digital records [18].

Table S5. Number of oil and gas wells drilled

Years	Oil	Gas	Of Oil & Gas Only % Oil	Of Oil & Gas Only % Gas	Source
1859-1956*	172750	11720	94%	6%	Table 1, Dilmore et al. [18]
1859-1928$^{\dagger\ddagger\diamond}$	134575	3859	97%	3%	Proved Fields, P. 63, Arnold & Kemnitzer [24]
1859-1928$^{\dagger\ddagger\diamond}$	178	120	60%	40%	Wildcat, P. 63, Arnold & Kemnitzer [24]
1929$^{\dagger\ddagger\diamond}$	2808	770	78%	22%	Scaled from Arnold & Kemnitzer [24]
1929-1956	3059	3857	44%	56%	Incl. Injection, Table SI-6 & SI-5, Dilmore et al. [18]
1929-1956	2026	3857	34%	66%	Excl. Injection, Table SI-5, Dilmore et al. [18]
1929-1956	37995	7723	83%	17%	Table SI-8, Dilmore et al. [18]
1930-1937‡	21618	10243	68%	32%	Scaled from Arnold & Kemnitzer [24] and PA DER Progress Reports [22]
1938-1949‡	21752	10307	68%	32%	Scaled from Arnold & Kemnitzer [24] and PA DER Progress Reports [22]
1930-1937$^{\dagger\diamond}$	14092	17769	44%	56%	Excl. Injection, Table SI-5, Dilmore et al. [18]
1938-1949$^{\dagger\diamond}$	14180	17879	44%	56%	Excl. Injection, Table SI-5, Dilmore et al. [18]
1950-1956$^{\dagger\ddagger\diamond}$	1160	9283	11%	89%	PA DER Progress Reports [22]
1957-2012*$^{\dagger\ddagger\diamond}$	38732	63010	38%	62%	Table 1, Dilmore et al. [18]
1950-1991	18890	21640	47%	53%	PA DER Progress Reports [22]
1992-2013	646	1339	33%	67%	PA DEP Oil and Gas Reports [1]
1859-2012	211482	74730	74%	26%	Table 1, Dilmore et al. [18]*
1859-2012	205726	112690	65%	35%	Our Estimate 1†
1859-2012	263903	113029	70%	30%	Our Estimate 2‡
1859-2012	357379	112690	76%	24%	Our Estimate 3$^{\diamond}$

Note that dry and other well types are not included in the above numbers.
* Numbers used in estimate by Dilmore et al. [18].
† Numbers used in our Estimate 1.
‡ Numbers used in our Estimate 2.
$^{\diamond}$ Numbers used in our Estimate 3, which includes a factor of 2.0 for oil wells drilled prior to 1938.